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Evidence of “crossed” transitions in dots-in-a-well structures through waveguide absorption measurements

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In-plane absorption measurements were performed at room temperature by means of a waveguide transmission setup on a Stranski–Krastanov InAs dots-in-a-well system emitting at 1.3 μm embedded in a *p-i-n* structure. The polarization dependence of quantum dot (QD) absorption was exploited to resolve its discrete and continuous spectral components and study them separately under reverse bias application. The quantum confined Stark effect observed in the discrete spectral component gave evidence of an upward built-in QD dipole of about 9.5×10^{-29} C m. The continuous component was found to originate from electronic transitions involving a QD state and a quantum well state. © *2008 American Institute of Physics*. [DOI: 10.1063/1.3000381]

InAs/InGaAs/GaAs Stranski–Krastanov dots-in-a-well layers grown by molecular beam epitaxy (MBE) have recently proved to be highly efficient as an active medium for semiconductor lasers at 1.3 μm, yielding high device performances such as high gain, high temperature stability, low threshold current density,[1–3] and high modulation bandwidth.[4,5] This suggests that effective capture and relaxation processes of the high-energy charge carriers towards the lasing ground state of the quantum dot (QD) must occur in these systems. Yet, in the most traditional picture of strictly discrete QD eigenstates, momentum conservation laws and selection rules lead to theoretical predictions which fail to justify an efficient intradot carrier relaxation.

Since its first experimental observation, in 1999,[6] the continuous background overlapping the discrete absorption spectrum of Stranski–Krastanov QDs, often referred to as the *continuum* (of electronic states), has been the subject of a few interesting theoretical and experimental studies;[7,8] in addition the account of such continuum in the QD carrier dynamics has provided further understanding of the intraband carrier relaxation.[9,10]

The nature of the absorption transitions forming the smooth spectral component is still a subject of debate, and a clear picture of the role played by the continuum in the intradot carrier dynamics is still missing primarily due to the lack of methods to directly probe QD absorption. Most of QD absorption measurements in literature, including those evidencing the presence of the continuum, are based on indirect techniques such as photoluminescence excitation, photocurrent detection, and electromodulated transmission.[6,8,11–14]

This letter addresses the experimental study of the QD continuum through direct optical transmission measurements at room temperature. To this aim, an optical transmission technique employing waveguide planar propagation of light was developed. The dependence on the optical polarization of QD optoelectronic properties was exploited to distinguish and separately measure the sole absorption continuum and the discrete states, whose selection rules were probed by applying an external reverse bias.

Figure 1(a) provides a schematic representation of the basic experimental setup used in this work. A *planar* transmission configuration was chosen to increase the interaction length and make the absorption detectable. Such configuration, coupled to a confocal microscope arrangement for collection and spatial filtering of light, has been already demonstrated to provide an effective way to measure QD absorption.[15] The optical setup was connected to an electrical probe station to allow electrical pumping and bias-dependent transmission measurements. A rotating polarizing filter was positioned on the optical axis in the infinity zone behind the microscope objective to probe the polarization dependence of the investigated systems.

The sample employed in this work is a single dots-in-a-well layer, full ridge laser structure grown by solid-source MBE [see details in Figs. 1(b) and 1(c)]. More details concerning the QD growth and the laser fabrication can be found in Refs. 2 and 16. A reference ridge sample without a dots-in-a-well layer was used to characterize the bare waveguide effects on the spectral transmission of the full structure.

The edge photoluminescence of the samples (not shown here) turns out to be polarized mostly along the direction parallel to the growth plane (hereafter referred to as “hori-

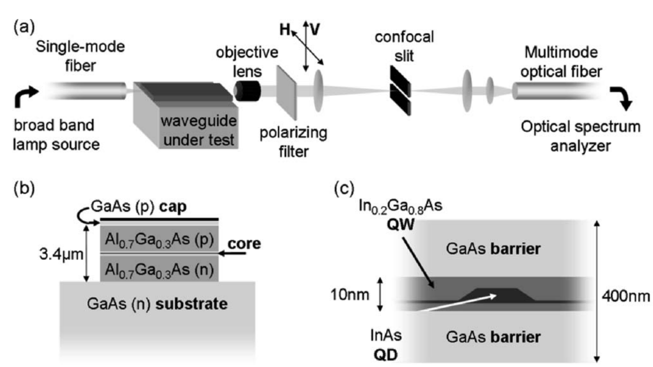

FIG. 1. (a) A scheme of the employed absorption setup. (b) Schematic representation of the investigated laser structures. (c) Details of the core region.

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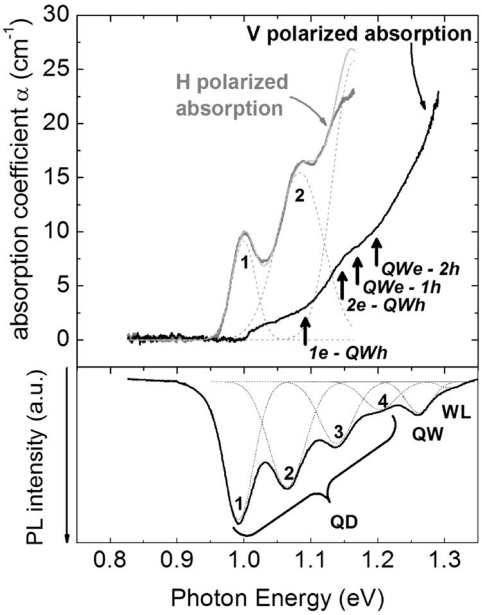

FIG. 2. Polarization-dependent absorption spectra (upper plot) and PL spectrum recorded at 300 K at a high excitation density (lower plot). The estimated onsets of the lowest energy bands of crossed transitions are indicatively shown by arrows (to be compared with Fig. 4); 0D confined transitions are numbered from 1 to 4.

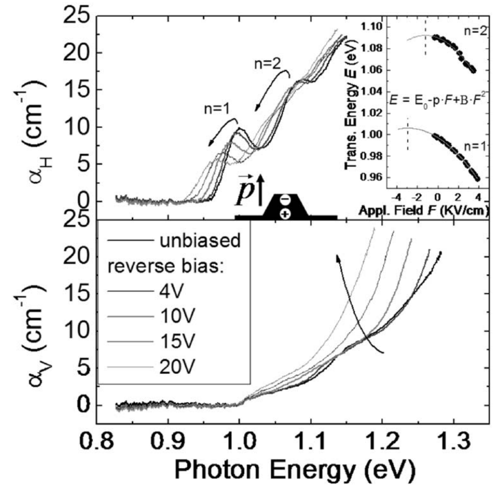

FIG. 3. Absorption spectra as a function of the reverse applied bias for both states of polarization. The inset in the upper plot shows the energy shift for the $n=1$ and $n=2$ transitions.

zontal" and labeled H), whereas a faint emission can be collected for the polarization perpendicular to the growth plane ("vertical," labeled V). Such behavior, which is observed to hold under electrical pumping as well (both below and above lasing threshold), is due to the dominant heavy-hole-like character of the QD radiative transitions.[12] Such a strong polarization dependence of the confined QD transitions is exploited to remove from the absorption spectrum their related features by using a polarizing filter, thus isolating the continuous absorption background.

In the upper plot of Fig. 2 the absorption spectra for both polarization states are presented, as calculated from

$$\alpha(h\nu) = -\frac{1}{L}\ln\left[\frac{I_{\mathrm{QD}}(h\nu)}{I_{\mathrm{ref}}(h\nu)}\right], \tag{1}$$

where $I_{\mathrm{QD}}(h\nu)$ and $I_{\mathrm{ref}}(h\nu)$ are the measured spectra after transmission through the waveguide with (QD) and without dots (ref) and L is the waveguide length. As mentioned above, the discrete QD absorption transitions, those involving zero-dimensional (0D) confined states, contribute only to the H-polarization spectrum $[\alpha_H(h\nu)]$. Just the ground state ($n=1$) and the first excited state ($n=2$) transitions could be detected, the higher energy absorption being too strong to allow light transmission. A multi-Gaussian fit provides a ground state absorption of 8.9 cm^{-1}, peaked at 999.6 meV with a spectral width $W=35.7$ meV. The measured Stokes shift (SS) with respect to PL (lower plot of Fig. 2) is around 10 meV, which is in good agreement with the expected value of 9.2 meV, as estimated by the formula SS $=0.18\ W^2/k_B T_C$, valid for an inhomogeneous QD ensemble in conditions of efficient carrier thermal escape,[17] at T_C $=290$ K.

In the V-polarization absorption spectrum $[\alpha_V(h\nu)]$ the discrete 0D features are suppressed. It shows just a smooth profile, which follows the intensity of the QD absorption background observed in similar systems through indirect experimental techniques.

According to Vasanelli *et al.*,[7] such QD continuous absorption background should be attributed to a continuum of transitions between discrete 0D confined states and two-dimensional (2D) states lying in a continuous energy band. Such transitions were predicted to arise from the partial overlap of carrier wavefunctions centered in different spatial regions, one in the QD and the other in the embedding material. As such, they are indirect in the real space and consequently often referred to as "crossed transitions" of the QD system. They contribute to the single QD absorption spectrum a monotonously growing background profile showing several rises related to the onset of different crossed transition bands.[7,8]

In our sample such changes in the slope of the continuum profile are less pronounced, as a consequence of the dispersion in size throughout the QD ensemble. Moreover, since the high-excitation PL shows a relatively intense peak at 1.262 eV related to the $\mathrm{In_{0.18}Ga_{0.82}As}$ quantum well (QW) radiative transition, it is reasonable to assume that the 2D states involved in the crossed transitions belong to the embedding QW structure. The onset energies of some of such QD-QW crossed transition bands (as obtained through a rough estimate whose details will be provided later in the text) are represented by arrows in the α_V plot of Fig. 2 for indication. An experimental evidence of the continuum consisting of QD-QW crossed transitions can be obtained through bias-dependent optical transmission measurements. Upon reverse bias, the band structure of the p-i-n heterostructure is distorted in such a way that the energies of the 0D confined carriers are lowered and their wavefunctions drift and concentrate along opposite directions for the electron and the hole. This causes a redshift in the related spectral features and their decrease in intensity according to the quantum confined Stark effect (QCSE). On the other hand crossed transitions, being indirect in the real space, are expected to show a somehow different behavior in the occurrence of band distortion.

The bias-dependent measurements are reported in Fig. 3. The optical absorption in the H polarization shows a clear QCSE: the two 0D absorption peaks are both seen to shift,

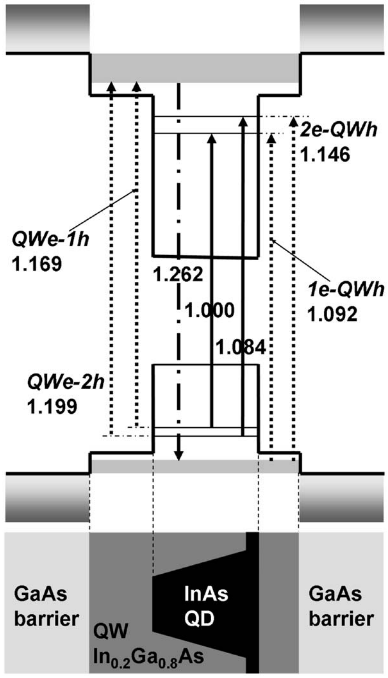

FIG. 4. Schematic representation of the transitions involved in QD absorption (labeled with their energy in eV). The dashed-dot line represents the QW ground state PL transition.

smear out, and weaken. In the inset of the upper graph of Fig. 3 the Stark shift of the two peaks is plotted versus F, the electric field generated in the intrinsic region, as an effect of the reverse bias application. By fitting the available data with the parabola $E=E_0-pF+BF^2$ (where p is the dipole moment), it is found that the dipole formed inside the QD by the displacement of the hole-electron couple is inverted (with respect to what is expected as an effect of the band distortion related to the built-in potential), showing the electron wavefunction closer to the top of the QD. This is not new in the literature,[13] and it can be justified by an interdiffusion of gallium and indium atoms during the growth, causing the formation of a potential gradient decreasing from the bottom up to the top of the QD. From the calculated dipole moments, an average distance of the $n=1$ ($n=2$) state wavefunctions of 5.94 ± 0.14 Å (3.75 ± 1.06 Å) was found, in good agreement with Ref. 13.

As for the bias dependence of the α_V spectrum, this is in fact remarkably different from α_H. In this case, absorption intensity increases with the reverse bias in the entire spectral range, whereas profile smoothness prevents ascertaining whether any spectral shift occurs. This is a confirmation of the real-space indirect nature of the transitions forming the continuum, since the applied reverse bias, while "pushing" the confined carrier wavefunctions towards the QD boundaries, causes a better overlap with the QW carrier wavefunctions to occur.

A rough estimate of the onset energies of such QD-QW crossed transition bands has been obtained by assuming that the In(Ga)As/GaAs band-offset ratio 0.65:0.35 (Ref. 7) is valid also to describe the relative energy displacements of the electron and hole levels from the respective band edges. Considering the energies derived from the spectra of α_H (for $n=1$ and $n=2$ QD transitions) and PL (for the QW ground state), a first crossed transition band, whose low-energy edge is estimated to be around 1.092 eV, is found to involve the $n=1$ electron state in the QD and the hole states in the QW (Ie-QWh, as represented in Fig. 4). The edge of this band, smeared by the QD ensemble dispersion, is responsible for the measured continuum onset, as can be qualitatively inferred from the upper plot of Fig. 2. Subsequently, the second and more intense absorption edge which is clearly seen above 1.1 eV is likely to correspond to the three following 0D-2D bands at higher energies (whose values are also reported in Fig. 4).

In conclusion, a waveguided planar transmission experiment in a dots-in-a-well system was carried out to study QD absorption. The polarization dependence of the spectra allowed studying separately the discrete QD spectrum and its overlapping continuous background. The intensity of the latter increasing under reverse bias constitutes an experimental demonstration that the QD continuum is formed by real-space indirect transitions involving the photogeneration of confined electrons (holes) in the QD and holes (electrons) in the embedding QW.

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